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Geochemical portray of the Pacific Ridge: New isotopic data and statistical techniques

Cédric Hamelin^{a,*}, Laure Dosso^b, Barry B. Hanan^{c,} Manuel Moreira^d, Andrew P. Kositsky^e & Marion Y. Thomas^e

^a I.U.E.M., U.B.O., Place Nicolas Copernic, 29280 Plouzané, France

^b Centre National de la Recherche Scientifique, UMR 6538, IFREMER, BP70, 29280 Plouzané, France

^c Department of Geological Sciences, S.D.S.U., 5500 Campanile Drive, San Diego, CA 92182-1020, USA

^d Institut de Physique du Globe de Paris, CNRS UMR 7154, 1 rue Jussieu, 75252 Paris CEDEX 05, France

^e Tectonics Observatory, California Institute of Technology, Pasadena, CA 91125, USA

* Corresponding author : Cédric Hamelin, I.P.G.P, 1 rue Jussieu, Bureau 345, 75252 Paris CEDEX 05, France. Tel.: +33 1 83 95 76 72; fax: +33 1 7193 7710, email address : hamelin@ipgp.fr

ABSTRACT:

Samples collected during the PACANTARCTIC 2 cruise fill a sampling gap from 53° to 41° S along the Pacific Antarctic Ridge (PAR). Analysis of Sr, Nd, Pb, Hf, and He isotope compositions of these new samples is shown together with published data from 66°S to 53°S and from the EPR. The recent advance in analytical mass spectrometry techniques generates a spectacular increase in the number of multidimensional isotopic data for oceanic basalts. Working with such multidimensional datasets generates a new approach for the data interpretation, preferably based on statistical analysis techniques.

Principal Component Analysis (PCA) is a powerful mathematical tool to study this type of datasets. The purpose of PCA is to reduce the number of dimensions by keeping only those characteristics that contribute most to its variance. Using this technique, it becomes possible to have a statistical picture of the geochemical variations along the entire Pacific Ridge from 70°S to 10°S. The incomplete sampling of the ridge led previously to the identification of a large-scale division of the south Pacific mantle at the latitude of Easter Island. The PCA method applied here to the completed dataset reveals a different geochemical profile. Along the Pacific Ridge, a large-scale bell-shaped variation with an extremum at about 38°S of latitude is interpreted as a progressive change in the geochemical characteristics of the depleted matrix of the mantle. This Pacific Isotopic Bump (PIB) is also noticeable in the He isotopic ratio along-axis variation. The linear correlation observed between He and heavy radiogenic isotopes, together with the result of the PCA calculation, suggests that the large-scale variation is unrelated to the plume–ridge interactions in the area and should rather be attributed to the partial melting of a marble-cake assemblage.

Research Highlights

New Sr, Nd, Pb, Hf, and He isotopes data fill a sampling gap along the Pacific Ridge. \blacktriangleright We examine geochemical variation in MORB using a principal component analysis. \triangleright A progressive change in the depleted matrix is recognized along the Pacific ridge. \triangleright In samples devoid of plume influence, He isotopes correlates with Pb isotopes.

Keywords: oceanic basalts; Pacific–Antarctic Ridge; mantle heterogeneity; Principal Component Analysis; Sr Nd Pb Hf isotopes

44 **1. Introduction**

45 Mid-oceanic ridge basalts (MORB) are the result of continuous melting of the ambient upper mantle beneath oceanic ridges. The MORB-source mantle is generally thought to have 46 been depleted ~2 Gy ago by extraction of the continents. Although the range of geochemical 47 48 variations in oceanic basalts is mostly attributed to the influence of Ocean Island Basalt 49 (OIB), significant geochemical heterogeneity in MORB has been recognized (Hoffman et al., 50 2003 and reference therein; Rudge et al., 2005). This heterogeneity has been revealed using 51 radiogenic isotope ratios (Sr, Nd, Pb and Hf) in areas devoid of plume influence. Numerous studies have attempted to model the mixing relationship between enriched and depleted 52 53 domains within the mantle to reproduce the range of isotopic variations observed in oceanic 54 basalts (Albarède, 2001; Meibom and Anderson, 2004; Rudge et al., 2005; Kellogg et al., 2007). The range of Sr, Nd and Pb isotopic compositions depends not only on the end-55 56 member compositions but also on the volume of mantle sampled during the melting relative to the length scale of heterogeneities (Kellogg et al., 2007). Therefore, the range of MORB 57 58 geochemical variations reflects the size, the spatial distribution, and the difference of 59 fusibility of heterogeneities within the upper mantle. In addition to this intrinsic heterogeneity of the MORB mantle, radiogenic isotope studies have led to the definition of distinct broad 60 61 mantle isotopic domains, such as the archetypal DUPAL anomaly located in the south 62 hemisphere (Dupré and Allègre, 1983; Castillo, 1988). Boundaries between these domains can be (i) extremely sharp as seen at the Antarctic Australian–Antarctic Discordance (ADD), 63 64 with isotopic ratios (Sr. Nd, Pb and Hf) abruptly changing from Indian to Pacific values (Pyle 65 et al., 1992, Hanan et al., 2004; Meyzen et al., 2007; Cooper et al., 2009) or (ii) more gradual 66 as seen in the transition from Southwest Indian Ridge to South Atlantic MORB (Meyzen et al, 67 2005).

68 Another example of these large geochemical provinces is given by the two sub-pacific 69 mantle domains (Vlastélic et al., 1999). The distinctive isotopic properties of these large-scale 70 geochemical domains suggest a long-term isolation of these mantle provinces, each with their own convective histories involving various amounts of melting residues and recycled 71 72 components. On the basis of an incomplete dataset, a boundary between these two pacific 73 provinces has been located at the latitude of the Easter Island microplate (Vlastélic et al., 74 1999). But in order to have a complete geochemical view of the southern Pacific Ridge from 75 66 to 10°S, it was necessary to fill the sampling gap between 53°S and 41°S. This became one 76 of the main objectives of the PACANTARCTIC2 cruise which took place in 2004-05 (Dosso 77 et al. 2005, Klingelhoefer et al., 2006, Moreira et al., 2008; Hamelin et al., 2010). Analyses of 78 Sr, Nd, Pb, Hf and He isotopic compositions of these new samples from the Pacific Antarctic 79 Ridge (PAR) are compiled here together with published data from 66°S to 53°S and from the 80 East Pacific Rise (EPR). Therefore it becomes possible to have a picture of the geochemical 81 variations along the entire Pacific Ridge from 66°S to the 10°S. Adapting a Principal 82 Component Analysis (PCA) to incomplete datasets, we show a detailed portray of the 83 geochemical variability of the Pacific Ridge.

84

2. Data selection and analytical methods.

86 2.1. The Pacific Ridge database from 10 to 66°S.

Along the Pacific Ridge, the Chile Triple Junction at 35°S/110°W separates the PAR from the EPR. The two pacific ridges show different geological settings: the full spreading rate increases along the PAR from 54 mm/yr at 65°S to 100 mm/yr at 35°S whereas it decreases along the EPR from 158 mm/yr at 35°S to 146 mm/yr at 10°S. In conjunction, the ridge axis morphology changes from a valley to a dome north of 60°S along the PAR (Ondréas et al., 92 2001), whereas the EPR is characterized by a uniform dome shaped morphology. Three 93 plume-ridge interactions generate abnormal morphological structures along this section: 94 Foundation (~38°S), Easter Island (~26°S) and 17°S. They are recognized by a high ridge 95 cross-sectional area (Klingelhöfer et al., 2006) and the presence of intense off-axis volcanic 96 activity. Compared to the EPR, the Pacific Antarctic Ridge still remains geochemically poorly 97 known. A large portion of this plate boundary has been previously surveyed (Lonsdale, 1994; 98 Cande et al., 1995) but the northern part has only recently been sampled (Dosso et al., 2005; 99 Moreira et al., 2008; Hamelin et al., 2010).

100

101 To generate a coherent database along the pacific ridges, on-axis samples analyzed for one 102 or more isotopic ratios (Sr-Nd-Pb-Hf-He) are selected from 66°S to 10°S using our new 103 isotope data (table 1) completed with the petrological database of the Lamont-Doherty Earth 104 Observatory (http://www.petdb.org). In most cases when the reference values of the standards 105 are reported, the data are normalized to the values of NBS987, 0.71025 for Sr, of JNdi-1, 0.512104 and La Jolla, 0.511852 for Nd, NBS981, ²⁰⁶Pb/²⁰⁴Pb=16.9373, ²⁰⁷Pb/²⁰⁴Pb=15.4925, 106 ²⁰⁸Pb/²⁰⁴Pb=36.7054, for Pb and JMC475, 0.282162 for Hf (see http://georem.mpch-107 108 mainz.gwdg.de/ for details and references).:

109

110 2.2. Analytical methods.

111 2.2.1. Double Spike Pb analyses along the PAR

New high-resolution Pb analyses were carried out for samples collected during PACANTARCTIC1 (PAC1) and PACANTARCTIC2 (PAC2) cruises along PAR segments from 66°S to 56°S and from 53°S to 41°S respectively (Fig. 1, Table 1). Small chips from the inner part of the pillow lavas were handpicked to avoid altered surfaces that could be a potential source of Pb contamination. Powdered samples were leached with 6M HCl at 140°C 117 for an hour and then rinsed up to 6 times with ultrapure water prior to dissolution. Lead 118 separation was then performed on an anionic exchange resin. Pb analyses were performed at 119 Ifremer (Centre de Brest) on a Finnigan MAT 26x multicollector instrument (MAT261 120 upgraded by Spectromat), using the double spike technique with the calibrated Southampton-121 Brest 207/204 spike (Ishizuka et al., 2003). Replicate analyses of the Pb isotope standard NBS981 gave an average of 16.9432±0.0027 and 15.5004±0.0029 and 36.7326±0.0086 for 122 ²⁰⁶Pb/²⁰⁴Pb, ²⁰⁷Pb/²⁰⁴Pb and ²⁰⁸Pb/²⁰⁴Pb, respectively. Pb blanks measured using this 123 124 procedure were < 100 pg, and thus negligible relative to the amount of sample analyzed.

125

126 2.2.2. Hf measurements along the PAR

Hafnium isotopic compositions were analyzed along the PAC1 ridge segments at SDSU on splits from the same samples (Hamelin et al., 2010). Hf was separated using the protocol of Blichert-Toft et al. 1997 with a negligible blank of less than 25 pg. Hf isotope ratios were measured at SDSU using the Nu Plasma. The ¹⁷⁶Hf/¹⁷⁷Hf was normalized for mass fractionation relative to ¹⁷⁹Hf/¹⁷⁷Hf=0.7325. The JMC-475 Hf standard ¹⁷⁶Hf/¹⁷⁷Hf gave 0.282160 \pm 0.000010 (2 σ) during this study. The standard was run alternately with samples to monitor machine performance.

134

135 2.2.3. He measurements along the PAR

Helium isotopic compositions have been measured on PAC1 and PAC2 samples at IPGP (Institut de Physique du Globe de Paris). Fresh pieces of glass were cleaned with distilled water, ethanol and acetone using an ultrasonic bath. Some samples were also cleaned with hydrogen peroxide in order to remove some Mg crust. Analytical procedure is identical to previous studies from our laboratory and can be found in Moreira et al. (1995). Samples were crushed under vacuum with analytical blanks of 7 ± 1 nccSTP ⁴He. This corresponds to 0.02 to 142 0.4% of the samples. Helium concentrations and isotopic composition were measured using
143 the ARESIBOII mass spectrometer (Moreira et al., 2008).

144

145 **3. New results compared to published MORB data along this**

- 146 ridge section.
- 147

These results include samples from 66°S to 56°S (PAC1) and from 53°S to 41°S (PAC2) (Fig. 1). New high resolution Pb analyses as well as Hf and He analyses are presented here in Table 1. The Sr, Nd and Hf analyses on the same samples are found in Vlastélic et al. (2000) and Hamelin et al. (2010).

152

153 3.1. Binary isotopic correlations

154 With 7 isotopic ratios, the number of possible binary isotope diagrams is 21. As an 155 example, we choose here to describe 4 such binary plots, selecting some of the most 156 commonly discussed (Fig. 2A-D). In all diagrams, pacific MORB samples devoid of plume 157 influence define linear correlations that in most cases have been previously described in the 158 literature. This result is in good agreement with the expected coherence in behavior between 159 Rb-Sr, U-Pb, Th-Pb, Sm-Nd and Lu-Hf isotopic systems compared to each other during 160 magmatic processes. When plume-ridge interaction samples are included in our dataset, they define elongated fields overlapping PAR-EPR array except in Figure 2C where He- \mathcal{E}_{Nd} 161 162 isotopic ratios define sub-parallel trends which emerge from the PAR-EPR array and point towards higher ³He/⁴He (R/Ra) ratios. More than 30 years of He isotopes systematic of 163 164 oceanic basalts worldwide have shown that MORB samples are characterized by a narrow

range of composition compared to OIB samples. However even within this limited range, our data show a linear correlation between ${}^{3}\text{He}/{}^{4}\text{He}$ (R/Ra) values and \mathcal{E}_{Nd} (Fig. 2C). The correlation between He and other isotopic systems will be discussed further below (§4.4.2).

168 In contrast with the decoupling of Hf and Nd isotope compositions previously documented 169 in MORB (e.g., Patchett and Tatsumoto, 1980; Salters and White, 1998; Chauvel et al., 2001; 170 Debaille et al., 2006), a rather good correlation between these two isotopic systems is 171 observed along our studied are (Fig. 2B). Debaille et al. (2006) have suggested that a 172 distinctive behavior of Hf during disequilibrium melting along their studied ride area (Atlantic ridge 22-35°N) could explain a decoupled behavior of ¹⁷⁶Hf/¹⁷⁷Hf with respect to other 173 174 isotopic ratios. The linear correlations observed in our dataset, show that the hypothesis of a 175 specific behavior of Hf does not apply along the pacific ridges. Recently, good correlations 176 between Hf and Nd isotopic ratios have been reported from other ridge segments such as 177 Mohns Ridge (Blichert-Toft et al., 2005) and the entire mid-Atlantic Ridge (Agranier et al., 2005). 178

179

180 *3.2. MORB variability along the south Pacific Ridge.*

181 Latitudinal isotopic variations of south Pacific Ridge basalts are shown in Figure 3. An 182 important geochemical feature from 66 to 10°S is a large scale (spanning approximately 183 5000km) and coherent variation of all isotopes shown by a bell shape grey line. The 184 extremum of this variation defining the Pacific Isotopic Bump (PIB), is located at the latitude 185 of Foundation (38°S). It reveals a less depleted component, characterized by more radiogenic 186 values for Sr, Pb isotopic ratios, and correlatively less radiogenic values for Nd and Hf. Three 187 shorter wavelength variations of the order of 200 to 700km are seen as isotopic anomalies 188 superimposed on the otherwise bell shaped curve of the isotope signature along the ridge. In a 189 way similar to the Pacific Isotopic Bump, these variations are towards more radiogenic Sr and Pb values and coherently less radiogenic Nd and Hf values. They indicate the influence ofenriched materials due to plume-ridge interactions.

192 Helium isotopes do not fit this Sr-Pb/Hf-Nd coherent behavior. At the latitude of the Foundation-Ridge intersection (38°S), the bell-shaped He curve shows radiogenic ³He/⁴He 193 194 ratios. But on the other hand, hot-spot influenced samples form negative "plume anomalies" with very unradiogenic signatures (high ${}^{3}\text{He}/{}^{4}\text{He}$ ratios, up to 12 R/Ra). The amplitudes of the 195 196 PIB and the anomalies attributed to the plume effect are of the same order of magnitude for 197 Nd, Hf and Pb. For He and Sr, the PIB has much smaller amplitude than the hotspot 198 anomalies. Additionally, local MORB variability is expressed as spikes, which are likely 199 related to transform faults (Eltanin System).

200

201 **4. Discussion**

202 4.1 Statistical definition of mantle reference lines

203 In most binary plots of radiogenic isotopic ratios, representative points of mantle derived 204 material define linear trends. Historically, these trends have been used to define reference 205 lines such as the Northern Hemisphere Reference Line (NHRL) in Pb-Pb plot (Hart, 1984) or the "Mantle Array" in Sr-Nd plot (DePaolo and Wasserburg, 1979) and the Nd-Hf plot 206 207 (Vervoort and Blichert-Toft, 1999). These reference lines are arbitrary and depend on the abundance of data available at the time of their definition. However, the data colinearity in 208 209 these plots justifies these convenient choices. The NHRL has been convenient to quantify the 210 Dupal anomaly using the $\Delta 7/4$ and $\Delta 8/4$ (Hart, 1984). In 1999, new MORB data from the 211 south Pacific lead to the definition of the Pacific Reference Line (PRL) which proved 212 convenient to compare two sub-pacific mantle domains, using the δ (Nd-Sr) and δ (Sr-Pb) 213 notations (Vlastélic et al., 1999). With the recent advance in analytical MC-ICPMS

214 techniques, the number of isotopic data increases dramatically. It becomes possible to look at 215 these reference lines in multidimensional space from a statistical point of view. Principal 216 Component Analysis (PCA) is a powerful mathematical tool to study data sets such as a 217 geochemical database including Sr-Nd-Pb-Hf analyses. The purpose of PCA analysis is to 218 reduce the number of dimensions in a data set by keeping those characteristics that contribute 219 most to its variance. This technique has been used in previous mantle heterogeneity studies 220 (e.g. Agranier et al., 2005, Debaille et al., 2006) The PCA method used with our geochemical 221 dataset has been initially developed for low-rank matrix approximations (Srebro and Jaakkola, 222 2003) and was recently adapted for tectonic problems using incomplete geodetic times series 223 (Kositsky and Avouac, 2010). The main difference with traditional PCA methods is that the 224 singular value decomposition is replaced by a more sophisticated decomposition, which 225 appropriately takes into account samples with a missing isotope measurement. This technique 226 is particularly suitable to geochemical data as it allows computing the principal components 227 using the whole dataset, increasing therefore the accuracy of the calculation (see 228 supplementary material for a more detailed discussion about the concept and the limits of our 229 calculation). Although more recently ICA (Independent Component Analysis) has been 230 chosen by some authors (Iwamori and Albarède, 2008, Iwamori et al., 2010), the preferred 231 PCA method used here has the advantage of dealing with missing isotopic data, assuming that 232 decorrelation is still a good assumption of independence in our dataset.

Because the PCA method is an orthogonal linear transformation, it assumes the linearity of the data co-variations. In most binary isotopic diagrams, mixing processes are expressed by hyperboles whose curvatures depend on the elemental concentration ratios of the involved end-members. But in the case of MORB, pseudo-linear correlations are observed (Fig. 2) indicating that denominator elements are in approximately constant proportions in the mixing components. It is very difficult to evaluate the extent of non-linear relationships concealed 239 within the analytical noise. However, it is worth noting that the geographical variations of the 240 components calculated with our method are approximately the same as those computed in the 241 3-dimensional space of Pb isotopes, in which relationships are linear. In order to minimize the correlations induced by the ²⁰⁴Pb analytical noise (it represents only about 1.4% of the total 242 Pb), the computation has been made in the ²⁰⁴Pb/²⁰⁶Pb, ²⁰⁷Pb/²⁰⁶Pb and ²⁰⁸Pb/²⁰⁶Pb space. 243 244 Considering the problematic of our study, this observation suggests that the curvature can be 245 neglected. One drawback of PCA in general stems from the fact that PCA is a projection 246 method, and sometimes low-dimensional visualization can lead to erroneous interpretations.

247 *4.2. Distribution of the variance among the principal components.*

248 The most striking result of the PCA is that the first principal component accounts for more 249 than 70% of the total variance. This result is remarkable considering the number of 250 geochemical parameters involved in the very large number of samples used in this analysis. It 251 indicates the strong coherence of all isotopic ratios within the depleted mantle domain. These 252 correlations in binary plots of isotopic systems are the result of two antagonistic processes 253 that took place over time: chemical fractionation events leading to the existence of enriched 254 and depleted mantle reservoirs and mechanical mixing of theses reservoirs during mantle 255 convection. The chemical fractionation between parent and daughter isotopes is controlled by 256 distribution coefficients. Even if these coefficients can be modified by multiple parameters, 257 they remain consistent from one element to another. Since these processes are approximately 258 linear, the resulting dispersion is located along a line corresponding to PC1 (the first Principal 259 Component).

260 While the importance of the first component is indisputable, one challenge with PCA is to 261 establish the number of significant components needed to explain the observed data variance. 262 A classical way to illustrate the number of relevant components is to study the residual 263 variance (χ^2) obtained for each component (e.g. Kositsky and Avouac, 2010). This value 264 drops abruptly after the third component (see supplementary material). At first, we can infer 265 that the information brought by the fourth to the sixth principal components is statistically 266 insignificant and can safely be ignored. Using a PCA computed in the 3-dimensional space of 267 Pb isotopes, the principal components account for 91.7%, 8.2% and 0.1% of the variance for 268 PC1, PC2 and PC3 respectively. This observation suggests that the variance expressed by 269 principal components over the order of 2 is the result of a randomly distributed error. Also the 270 absence of coherent geographical variation for the third component suggests that this variance 271 could either be a very local variation or an artifact of our data compilation (i.e. sampling bias, 272 data and error normalization between different laboratories). This justifies to limit the 273 discussion of the south pacific mantle heterogeneity to the PC1 and PC2 characteristics.

274

4.3. Mapping geochemical heterogeneities in the mantle using a PCA method.

276 Using the dataset available at the time, Vlastélic et al. (1999) identified a large geochemical variation in the south Pacific depleted mantle. Based on an incomplete sampling 277 278 of the ridge, they suggested the existence of a sharp boundary located at the latitude of the 279 Easter Island Microplate (27°S). As an interpretation, they proposed that the Pacific 280 Superswell divided the mantle into two domains each with their own convective histories, 281 producing slight differences in their average isotopic signatures. More recently, the origin of 282 these two domains has been challenged by a model based on plate kinetics (Small and 283 Danyushevsky, 2003). Small and Danyushevsky proposed that geochemical discontinuities 284 result from variations of the asthenosphere consumption, which corresponds to the ratio 285 between the accretion rate and the spreading center migration relative to plumes. Their model 286 predicted that the fast spreading, slowly migrating East Pacific Rise should have higher 287 average melting degrees compared to the slower spreading, rapidly migrating Pacific-288 Antarctic Rise. In order to identify the two mantle domains along the Pacific Ridge, Vlastélic

289 et al. (1999) used δ (Sr-Pb) and δ (Nd-Sr), which are defined as the vertical deviations from 290 references lines in Pb vs Sr and Sr vs Nd isotopic ratio plots respectively. These reference 291 lines (see §4.1) were drawn intuitively in the greatest variance of their dataset. They can be 292 directly compared to our first principal component. By construction, whatever the number of 293 dimensions considered in the PCA, the projection of PC1 in a binary diagram resembles the 294 relevant reference line (Fig. 2). Because PC2 is orthogonal to the greatest variance, sample 295 values along this component are correlated to delta notations such as $\delta(Sr-Pb)$, $\delta(Nd-Sr)$ or 296 Δ 8/4Pb. But compared to delta notations, PC2 has the advantage of being rigorously and 297 statistically determined in a multidimensional isotopic space. The application of the PCA 298 method to our data compilation (excluding He isotopes) reveals a geochemical profile of the 299 Pacific Ridge. The plot of PC1 versus latitude summarizes all the characteristics noted with 300 the different isotopic systems (Fig. 4). The short scale geochemical variations associated with 301 hotspots as well as the large-scale variation are clearly visible. 17°S and 25°S (Easter Island) 302 are well defined by sharp anomalies superimposed on the bell shaped curve. A plot of PC2 303 versus latitude shows a very different picture (Fig. 4): only the large scale variation is 304 expressed by this component, the geochemical variations associated with hotspots are 305 flattened and the corresponding samples are projected along the bell shape curve defining the 306 Pacific Isotopic Bump. This PIB is also shown on plots of delta versus latitude, illustrating the 307 equivalence of these parameters and PC2 (Fig. 4). At the Juan Fernandez/Foundation latitude 308 (36°S), the isotopic variation curve reaches an extreme which corresponds to a less "depleted" 309 isotopic signature. In contrast with the conclusion of Vlastélic et al. (1999), we proposed that 310 geochemical variations along the Pacific Ridge are not the result of two separated mantle 311 domains but should rather be seen as a progressive variation of the isotopic composition of the 312 sub-Pacific depleted mantle. Since the asthenosphere consumption varies abruptly at the Chile 313 triple junction and is almost constant along the PAR, the plate kinematic model of Small and

314 Danyushevsky (2003) is also inconsistent with the observed progressive variation of the315 MORB depleted matrix.

316

4.4. Mixing relationship within the depleted mantle compared to ridge-hotspotinteractions.

319 4.4.1. Asthenospheric versus hotspots signals as illustrated by Sr-Nd-Pb-Hf.

320 Except in figures involving He isotopes, geochemical variations related to ridge/hotspot 321 interactions are consistent with variations of samples devoid of plume influence. At first, it is difficult to distinguish the two types of variations in binary isotopic plots (Fig. 2). 322 Nevertheless, a PCA computed with all heavy radiogenic isotopes clearly illustrate a 323 324 difference: hotspot signatures are exclusively visible with PC1, while the variance related to 325 the large scale variation (PIB) is illustrated by both PC1 and PC2 versus latitude (Fig. 4). In 326 PC1 versus PC2 space, the MORB field extends from the depleted end member of the mantle (DMM) toward a recycled oceanic crust end member with a HIMU affinity (Fig. 5). The 327 328 samples identified as resulting from a plume-ridge interaction are not part of this "depleted 329 trend". Rather their data field extends from the depleted trend towards more enriched-type 330 end members such as C and/or EM. This observation supports the idea that the large scale 331 variation in the depleted Pacific mantle is unrelated to ridge/hotspot interactions. This 332 variation is therefore equivalent to the intrinsic variability of MORB recognized in other areas 333 (e.g. Dosso et al., 1999, Donnelly et al., 2004; Debaille et al., 2006; Hémond et al., 2006). It 334 has to be noted that PCA calculations assume a linearity criteria which is not satisfied when 335 dealing with isotopic compositions of the mantle end-members. It is thus not possible to 336 determine more precisely the nature of the different mantle components responsible for the observed trends. Nevertheless, the PCA calculations establish that the progressive 337

338 geochemical change of the depleted matrix of the Pacific mantle is not the result of hotspot 339 material being diluted into the depleted mantle. Using a different approach, Meyzen et al. 340 (2007) successfully unscrambled the hotspot and asthenospheric signals. These authors have 341 proposed that the geochemical variations unrelated to ridge/hotspot interactions along the 342 South West Indian ridge and the South Atlantic ridge are related to a broad lower mantle 343 upwelling in this area. Similarly, a broad lower mantle input could be a plausible cause of the 344 Pacific Isotopic Bump associated with the high spreading rates in the vicinity of the Chile 345 Ridge Triple junction.

346 It is interesting to compare the results of our PCA calculation along the Pacific ridge with 347 the spectral analysis performed along the Atlantic ridge by Agranier et al. (2005). These 348 authors found two contrasting types of spectra along their study area. The first type is 349 associated with ridge-hotspot interactions and is seen in the first principal component. The 350 second type is illustrated by the continuous power decrease with the decreasing wavelength of 351 the second principal component. Agranier et al. (2005) have interpreted this second type of 352 spectra, unrelated to hotspots, as being the result of the continuous size reduction of mantle 353 heterogeneities upon stretching and refolding of the convecting mantle. Despite the 354 differences in geological settings between the Pacific ridge and the Atlantic ridge, our 355 observations are in good agreement with the statistical analysis of Agranier et al. (2005). We 356 view the PIB as the consequence of a progressive change in the relative proportions of the 357 marble-cake components present in the Pacific upper mantle. In this model, hotspot anomalies 358 are superimposed on the intrinsic mantle heterogeneity expressed in MORB.

359

360 *4.4.2. He isotopes in the depleted mantle compared to hotspot signals.*

361 Unlike other isotopic systems, helium shows a first order discrepancy between the hotspot362 and the depleted mantle signals in our studied area (Fig. 3 and Fig. 6): the large scale variation

363 associated to the depleted mantle is characterized by an increase towards more radiogenic (low ³He/⁴He) compositions, whereas the short scale variation associated to the mantle plumes 364 points towards high ³He/⁴He values as previously noted in Hanan and Graham (1996). The 365 origin of the large range of elevated ³He/⁴He values in OIB lavas is a long-lived controversial 366 367 question (Kurz et al., 1982; Allègre et al., 1983; Meibom et al., 2003; Moreira et al., 2004). Historically, the requirement of a high ³He/⁴He reservoir for the OIB lead to the idea that 368 369 plumes are tapping a deep, undegassed lower mantle, isolated from the upper mantle 370 convection (O'Nions et al., 1996; Allègre et al., 1997). Numerous alternative models have tried to solve the apparent inconsistency between the high ³He/⁴He ratio in plumes and the 371 372 requirement of a source previously processed through partial melting (e.g. Parman et al., 373 2005; Class and Goldstein, 2005; Purtika, 2008; Albarède, 2008; Davies, 2010).

Compared to plume-influenced samples, systematics of He isotopes in depleted MORB 374 samples have shown a very restricted range of ${}^{3}\text{He}/{}^{4}\text{He}$ with a peak of distribution around 8 ± 1 375 376 (R/Ra) (Allègre et al., 1995). Within the uncontaminated depleted mantle sampled along our studied area, ³He/⁴He varies from 6 to 9.5 (R/Ra). Samples devoid of plume influence fall 377 along a negative trend toward low ³He/⁴He values and high ²⁰⁶Pb/²⁰⁴Pb. This variation of He 378 isotopes in our samples devoid of plume influence is clearly related to the PIB identified 379 380 along the pacific ridge using PCA with Sr, Nd, Pb and Hf isotopes. It is interesting to note that the most enriched samples from this correlation show the lowest ³He/⁴He values (more 381 radiogenic). In order to reconcile the ³He/⁴He variations with variations of Sr, Pb, Nd and Hf 382 383 isotopes, new models take into account the physical specificities of He compared to heavy 384 radiogenic isotopes: its diffusivity in mantle conditions and its capacity to be outgassed from 385 melts at sub-surface pressure (e.g. Hart et al., 2008; Albarède et al., 2008; Gonnermann and 386 Mukhopadhyay, 2009). We propose that the melting of a marble-cake upper mantle, 387 unpolluted by plumes, produces the good correlation observed between He and Pb isotopes in

our depleted samples. The less depleted component of the mantle assemblage is characterized by a high Pb isotopic ratio and a low ${}^{3}\text{He}/{}^{4}\text{He}$ ratio. We suggest that this component was derived from the recycling of an extensively outgassed oceanic crust. When the oceanic crust is subducted back into the mantle, it contains negligible concentration of mantle-derived He (Staudacher and Allègre, 1988). With time, this very low ${}^{3}\text{He}/(\text{U+Th})$ in subducted slabs is expected to produce the end-member with high Pb isotopes value and low ${}^{3}\text{He}/{}^{4}\text{He}$ ratio (Fig. 6).

395 In our model, the variation of He isotopes along the pacific ridge could be interpreted as: 396 (1) a smaller contribution of the refractory layers (less radiogenic), corresponding to a lower ${}^{3}\text{He}/{}^{4}\text{He}$ ratio at the PIB but because the melting rate is expected to be higher near the Chile 397 398 Ridge Triple Junction, this hypothesis seems very unlikely, or (2) a higher volume of the 399 recycled end-member in this area. This hypothesis is in agreement with the conclusions 400 derived from the PCA based on heavy radiogenic isotopes (§4.4.1). The intrinsic geochemical 401 variation of the depleted upper mantle comforts the image of a marble cake mantle composed 402 of a refractory component and a recycled oceanic crust component. A broad volume of 403 recycled component associated with the high spreading rates in the vicinity of the Chile Ridge 404 Triple junction is therefore a plausible cause of the Pacific Isotopic Bump. The correlation 405 between He and Pb isotopes revealed by our new data (Fig. 6) suggests that the depleted 406 mantle is the outcome of a mixing of a different nature that the one involved in the plume-407 ridge interaction.

408

410 **5. Conclusion**

411 Analyses of this new sampling of the PAR complete the dataset of the Pacific Ridge. 412 Therefore it becomes possible to have a picture of the geochemical variations from 10 to 413 70°S. Our data show a clear geographical evolution of isotopic characteristics along the 414 Pacific Ridge. In binary isotopic plots, this large-scale variation is expressed by correlations 415 between each isotopic dimension. This observation holds true even for the Nd-Hf system 416 previously reported as decoupled for MORB samples (Debaille et al., 2006). Even more significant in this study is the linear correlation shown in the isotopic He-Nd and ³He/⁴He -417 ²⁰⁶Pb/²⁰⁴Pb plots. In these isotopic spaces, the samples affected by plume-ridge interactions 418 419 depart clearly from the linear correlations displayed by the ridge samples coming from the 420 depleted mantle.

421 Despite the paucity of combined Sr-Nd-Pb-Hf isotope data on individual samples, the PCA 422 algorithms used here allow us to portray the variation along a huge section of the Pacific Ridge from a statistical point of view. Compared to previous studies of the sub-Pacific 423 424 mantle, the application of PCA reveals a Pacific Isotopic Bump, which can be seen as a 425 progressive geochemical variation of the depleted upper-mantle matrix rather than a sharp 426 frontier between two mantle domains. Combining PCA results with the information given by 427 He isotopes, we suggest that the Pacific Isotopic Bump is unrelated to plume-ridge 428 interactions. This geochemical variation in the upper mantle reservoir is the result of a 429 marble-cake mantle assemblage composed of a residual mantle component and a recycled 430 oceanic crust component.

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- 594

595 Figure captions

596

Figure 1: Map of the south pacific basin showing the location of the PACANTARCTIC1 and
PACANTARCTIC2 cruises with respect to the Pacific Antarctic Ridge (PAR) and the East
Pacific Rise (EPR). Hotspot locations are shown in orange on the map.

600

601 Figure 2: Examples of binary plots showing correlations within the MORB field. Samples 602 devoid of plume influence are shown in blue whereas samples from plume-ridge interactions 603 are in red. Blue circles represent samples collected along the PAR and blue crosses for EPR 604 samples (all symbols are kept the same in all figures). Mantle reference lines defined in the 605 literature are shown in dashed lines: in the Sr/Nd plot (A), the Pacific Reference Line (PRL) 606 (Vlastélic et al., 1999), in the Nd/Hf plot (B), the mantle array (Vervoort et al., 1999), in the 607 Pb/Pb plot (D), the North Hemisphere Reference Line (NHRL) (Hart, 1984). Since no 608 reference line had yet been recognized in plots involving He isotopes, we show here a regression line (${}^{3}\text{He}/{}^{4}\text{He} = 0.63 \epsilon_{Nd} + 1.74$, r = 0.89) (C). The projection of the two first 609 610 principal components (PC1 and PC2) calculated in our study are shown (solid black and grey 611 lines, see text for additional information). Data references can be found in supplementary 612 materials.

613

Figure 3: Geochemical variations along the PAR and the EPR from 66°S to 10°S. Dashed lines represent the location of major transform faults in the area and a grey shade is used for the Juan Fernandez and Easter Island microplates. The grey curve underlines the large-scale isotopic variation along the pacific ridges from 10 to 66°S and defines the Pacific Isotopic Bump (PIB).

620 **Figure 4:** Plot of the two first principal components along the Pacific Ridge. The similarity 621 between PC2 and δ notation is illustrated by plotting δ (Sr-Pb) as defined by Vlastélic et al. 622 (1999) and Δ^{207} Pb as defined by Hart (1984) versus latitude.

623

Figure 5: Plot of PC2 versus PC1 for a principal component analysis computed using Sr, Nd, Hf and Pb isotopes. The hashed blue field defines the depleted trend and includes EPR and PAR samples. Data points are drawn as ellipses representing the 95% confidence domain of the components as calculated in Debaille et al. (2006). The insert shows the location of the classical mantle end-members in the PC2 vs PC1 space.

629

Figure 6: Binary plot of ³He/⁴He vs ²⁰⁶Pb/²⁰⁴Pb. The dashed regression line (³He/⁴He = -2.33
²⁰⁶Pb/²⁰⁴Pb + 51.0) is defined by samples devoid of plume influence. Plume-ridge interactions
(17°S and Easter hotspots) are characterized by positive trends emerging from the PAR-EPR
array and pointing towards higher ³He/⁴He (R/Ra) ratios and higher ²⁰⁶Pb/²⁰⁴Pb.
Table 1: Complete Sr, Nd, Pb, Hf and He isotope data table for PACANTARCTIC 1 and 2

636 samples. In italic font, previously published data (Vlastélic et al., 1999; Vlastélic et al., 2000;
637 Moreira et al., 2008; Hamelin et al., 2010).













Table 1: Complete Sr, Nd, Pb, Hf and He isotope data table for PACANTARCTIC 1 and 2 samples. In italic font, previously published data (Vlastélic et al., 1999; Vlastélic et al., 2000; Moreira et al., 2008; Hamelin et al., 2010).

	⁸⁷ Sr/ ⁸⁶ Sr	¹⁴³ Nd/ ¹⁴⁴ Nd	8 _{Nd}	²⁰⁶ Pb/ ²⁰⁴ Pb	²⁰⁷ Pb/ ²⁰⁴ Pb	²⁰⁸ Pb/ ²⁰⁴ Pb	$^{176}\text{Hf}/^{177}\text{Hf}$	$\epsilon_{\rm Hf}$	³ He/ ⁴ He	Long. (°)	Lat. (°)	Depth
PACANTARCTIC 2												
PAC2DR38-1	0.702465	0.513108	9.17	18.671	15.533	38.039	0.283125	12.50	7.2	-111.3	-41.80	2524
PAC2DR37-1	0.702462	0.513091	8.84	18.722	15.540	38.110	0.283106	11.80	7.3	-111.3	-42.27	2475
PAC2DR37-2							0.283118	12.25	7.3	-111.3	-42.27	2475
PAC2DR36-1	0.702479	0.513066	8.34	18.724	15.538	38.117	0.283119	12.27	7.5	-111.6	-42.95	2503
PAC2DR35-1a	0 702402	0 512007	0.05	10 726	15 550	20 146	0.283121	12.34	7.2	-111.8	-43.59	2463
PAC2DR35-2 PAC2DP34-1	0.702492	0.513097	8.95 0.17	18./30	15.550	38.140	0.285127	12.54	7.2	-111.8	-45.59	2403
PAC2DR34-1 PAC2DR33-1	0.702488	0.513082	8.66	18.817	15.562	38.236	0.283111	11.99	7.26	-112.3	-44.87	2374
PAC2DR32-1	0.702516	0.513078	8.58	18.809	15.539	38.202	0.283100	11.60	7.2	-112.4	-45.39	2384
PAC2DR31-3	0.702479	0.513066	8.36	18.833	15.554	38.242	0.283112	12.03	7.3	-112.7	-45.85	2414
PAC2DR30-1	0.702472	0.513060	8.24	18.887	15.565	38.311	0.283106	11.83	6.8	-112.9	-46.40	2345
PAC2DR29-1	0.702504	0.513059	8.22	18.798	15.538	38.208	0.283117	12.19	7.2	-113.1	-47.01	2407
PAC2DR28-2	0.702468	0.513070	8.43	18.725	15.539	38.121	0.283102	11.66	7.1	-113.3	-47.51	2489
PAC2DR27-1	0.702643	0.513035	/./4	19.168	15.581	38.624	0.283094	11.3/	0.1 7.25	-113.4	-48.18	2359
PAC2DR22-1 PAC2DR22-3	0.702403	0.515100	9.01	18.720	15.559	38 202	0.283131	12.09	7.23	-113.0	-48.73 -48.73	2413
PAC2DR21-2	0.702483	0.513088	8.78	18.768	15.544	38.189	0.283125	12.48	7.1	-113.8	-49.26	2339
PAC2DR9-1	0.702499	0.513088	8.78	18.768	15.540	38.202	0.283122	12.37		-117.0	-49.59	2380
PAC2DR9-2				18.798	15.552	38.244	0.283132	12.73		-117.0	-49.59	2380
PAC2DR20-1	0.702493	0.513092	8.86	18.749	15.540	38.168	0.283126	12.51	7.1	-116.8	-49.73	2441
PAC2DR8-1	0.702483	0.513077	8.56	18.796	15.550	38.237	0.283132	12.74	7.3	-117.1	-49.99	2221
PAC2DR8-2	0 702 4 40	0.512000	0.70	18.670	15.535	38.031	0.283140	13.03	7.4	-117.1	-49.99	2221
PAC2DR/-4 PAC2DP6-1	0.702449	0.513088	0.70 8.00	18.741	15.528	38.148	0.285155	12.84	7.4	-117.2	-50.25	2229
PAC2DR6-6	0.702396	0.513094	8.99	18.792	15.543	38.123	0.283133	13.12		-117.2	-50.70	2610
PAC2DR5-2g	0.702524	0.513098	8.97	18.674	15.537	38.110	0.283135	12.85	7.5	-117.4	-50.98	2784
PAC2DR4-2	0.702367	0.513134	9.68	18.531	15.513	37.942	0.283163	13.83	7.7	-117.8	-51.43	2409
PAC2DR3-1							0.283139	12.97	7.5	-118.0	-51.79	2397
PAC2DR3-3	0.702382	0.513112	9.25	18.701	15.526	38.092	0.283143	13.12	7.5	-118.0	-51.79	2397
PAC2DR2	0.702417	0.513116	9.32	18.724	15.530	38.110	0.283141	13.06	76	-118.1	-52.13	2405
PAC2DRI-I	0.702422	0.313090	0.01	18.822	15.547	38.199	0.283133	12.75	7.0	-118.4	-52.55	2323
PACANIARCI	IC I	0 512120	0.75	19 404	15 490	29 100	0.202225	16.02	76	145.00	56.00	2617
PACIDRI4-2 PACIDRI4-3	0.702538	0.513158	9.75	18.494	15.469	38 379	0.283223	15.02	7.0	-145.09	-56.00	2617
PAC1DR14-4	0.702557	0.513132	9.62	18.769	15.523	38.370	0.283214	15.77		-145.09	-56.00	2617
PAC1DR13-2g	0.702556	0.513142	9.82	18.508	15.503	37.954	0.283204	15.28	7.6	-145.74	-56.57	2674
PAC1DR13-3	0.702570	0.513145	9.89	18.508	15.505	37.963	0.283200	15.14		-145.74	-56.57	2674
PAC1DR12-1g	0.702310	0.513150	9.99	18.064	15.467	37.468	0.283235	16.37		-146.29	-57.18	2539
PAC1DR12-3g	0.702375	0.513146	9.91	18.194	15.473	37.627	0.283217	15.74		-146.29	-57.18	2539
PACIDRI2-3r	0.702395	0.513170	10.38	18.192	15.470	37.620	0.283216	15.70		-146.29	-57.18	2539
PACIDRII-Ig PACIDR11-3	0.702455	0.513125	9.50	18.420	15.491	37.816	0.283182	14.11		-140.80 -146.80	-57.63	2500
PAC1DR10-1g	0.702403	0.513112	9.25	18.426	15.492	37.881	0.283149	13.33		-148.50	-57.89	2319
PAC1DR10-3	0.702470	0.513127	9.54	18.400	15.483	37.798	0.283148	13.30		-148.50	-57.89	2319
PAC1DR09-g	0.702467	0.513120	9.40	18.638	15.511	38.075	0.283150	13.37		-149.14	-58.85	2484
PAC1DR09-1	0.702372	0.513112	9.25	18.376	15.470	37.819				-149.14	-58.85	2484
PAC1DR08-2	0.702388	0.513110	9.21	18.406	15.480	37.827	0.283150	13.37		-150.02	-59.5	2365
PACIDR08-3	0.702376	0.513113	9.27	18.398	15.475	37.809	0.283149	13.33	7.8	-150.02	-59.5	2365
PAC1DR07-1g	0.702472	0.513099	0.99 8 07	18.620	15.505	38.059	0.283144	12.10		-152.08	-60.00	2362
PAC1DR07-2g	0.702434	0.513090	8.82	18.631	15.501	38.041	0.283122	12.03		-152.08	-60.00	2362
PAC1DR06-g	0.702502	0.513126	9.52	18.498	15.491	37.911	0.283163	13.83	8	-153.21	-60.94	2527
PAC1DR06-2	0.702389	0.513113	9.27	18.491	15.493	37.912	0.283171	14.11		-153.21	-60.94	2527
PAC1DR05-1g	0.702407	0.513132	9.64	18.489	15.495	37.926	0.283168	14,00		-154.54	-62.00	2344
PAC1DR05-3	0.702435	0.513138	9.75	18.490	15.496	37.936	0.283179	14.39	7.9	-154.54	-62.00	2344
PAC1DR05-r	0.702455	0.513122	9.44	18.482	15.488	37.904	0.283183	14.53	72	-154.54	-62.00	2344
PACIDR03-1 PAC1DR03-2	0.702421	0.513107	9.15	18.303	15.498	37.974	0.283132	13.44	7.5	-156.08	-62.52	2219
PAC1DR02-9	0.702335	0.513141	9.81	18.272	15.511	37.837	0.283189	14.75	1.5	-156.54	-62.64	2489
PAC1DR02-1	0.702362	0.513147	9.93	18.369	15.498	37.860	0.283186	14.64		-156.54	-62.64	2489
PAC1CV09	0.702370	0.513110	9.21	18.236	15.588	38.063	0.283139	12.98		-159.61	-62.66	2714
PAC1CV08	0.702419	0.513112	9.25	18.487	15.503	37.965	0.283126	12.52		-162.44	-62.77	2534
PAC1CV06-r	0.702362	0.513128	9.56	18.344	15.475	37.787	0.283169	14.04		-166.06	-63.45	2755
PACICV06-g	0.702397	0.513129	9.58	18.350	15.476	37.804	0.283172	14.15	0.1	-166.06	-63.45	2755
PACICV04-g	0.702512	0.513144	9.8/ 0.10	18.15/	15.462	31.313 37 052	0.2831/3	14.18 17 19	8.1	-109.40 -171.00	-04.4	2340 2576
PAC1CV03-g	0.702425	0.513119	9.34	18.492	15.486	37.923	0.283175	14.10		-171.88	-64.53	2576
PAC1CV02-g	0.702568	0.513135	9.69	18.391	15.485	37.868	0.283175	14.25	7.6	-172.43	-64.83	2936
PAC1CV01-r	0.702551	0.513153	10.05	18.447	15.557	38.121	0.283156	13.58		-173.75	-65.10	2863
PAC1CV01-g	0.702597	0.513124	9.48	18.761	15.591	38.549	0.283144	13.16		-173.75	-65.10	2863

Supplementary Material

New algorithm to compute a PCA:

The PCA method is a linear transformation that converts the data into a new coordinate system in which the direction along which the greatest data variance is expressed, becomes the first axis (PC1). The direction along which the greatest data variance is expressed orthogonal to PC1 becomes the second axis (PC2), and so on. The first step, called the whitening, consists in normalizing the data. This is done by subtracting the mean value from each data point and dividing the result by the standard deviation of each variable. The correlation matrix (Pearson's correlation matrix) of the reduced variables can then be calculated. The next step consists into diagonalizing the correlation matrix in order to find the eigenvalues, which correspond to the dimensions that have the strongest correlation in the data set. The principal components are the coordinates of the data points in the eigenvector referential.

The PCA method used with our geochemical dataset has been initially developed for low-rank matrix approximations (Srebro and Jaakkola, 2003) and was recently adapted for tectonic problems using incomplete geodetic times series (Kositsky and Avouac, 2010). The algorithm used to compute our principal components has been written specifically to address the major problem of a geochemical database: missing data in the matrix. Traditionally, for a given sample, it is necessary to acquire as many different isotope measurements as the number of dimensions used in the PCA. The paucity of combined Sr-Nd-Pb-Hf isotope data in individual samples is therefore an important issue for standard PCA algorithms. The more dimensions we use in the PCA, the fewer samples meet the required conditions: from 210 samples for 2-dimension (Sr-Nd) space to only 99 samples for a 6-dimension space. The main difference with a classical PCA is that we have replaced the standard Singular Value Decomposition (SVD) with a more sophisticated decomposition proposed by Srebro and Jaakkola [2003]. In this approach the data matrix X is decomposed into components U, S, V, with each data point weighted according to the square of its standard error. This decomposition is particularly adapted to take into account individual measurement errors and deal with missing data points. It allows us to compute the Principal Components on the entire data set, even if some isotopic ratios are missing. Each missing data point is weighted with infinite standard error. We want to emphasize that, consequently, no interpolation is made, which could erase some local effect. This technique is particularly suitable to geochemical data as it allows us to complete the principal components using the whole dataset, increasing therefore the accuracy of the calculation.

Limits of PCA:

Classical limits related to PCA calculation:

Because the PCA method is an orthogonal linear transformation, it assumes the linearity of the data co-variations. In most binary isotopic diagrams, mixing processes are expressed by hyperboles whose curvatures depend on the elemental concentration ratios of the involved end-members. But in the case of MORB, pseudo-linear correlations are observed (Fig. 2) indicating that denominator elements are in approximately constant proportions in the mixing components. It is very difficult to evaluate the extent of non-linear relationships concealed within the analytical noise. However, it is interesting to note that the geographical variations of the components calculated with our method are approximately the same as those computed in the 3-dimensional space of Pb isotopes, in which relationships are linear. In order to minimize the correlations induced by the ²⁰⁴Pb analytical noise (it represents only about 1.4% of the total Pb), the computation has been made in the ²⁰⁴Pb/²⁰⁶Pb, ²⁰⁷Pb/²⁰⁶Pb and ²⁰⁸Pb/²⁰⁶Pb space. Considering the problematic of our study, this observation suggests that the curvature can be neglected.

Limits related to our new algorithm:

Compared to traditional PCA algorithms, principal components are computed here altogether rather than separately, because the number of components affects the subspace in which these components reside. A limit to this method appears if one variable is represented by fewer samples than compared to other variables. In this particular case, a sample with a value corresponding to this "rare" variable is artificially given more weight. In return, each variable is also weighed in proportion to its number of samples. Because the number of He isotope analyses along the studied ridge section is too low compared to other isotopes, the information provided by this parameter has not been included in the PCA calculation. Distribution of the variance among the principal components:



Plot of the residual variance (χ^2) versus the principal component number

Figure 2 data references:

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